

Adsorption behavior of uranyl ions onto amino-type adsorbents prepared by radiation-induced graft copolymerization*

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Amino-type adsorbents (ATAs) were prepared by radiation-induced graft copolymerization of 4-hydroxybutyl acrylate glycidyl ether (HB) onto a polyethylene-coated polypropylene (PE/PP) duplex fiber of a non-woven fabric, and modified with different amines of ethylenediamine (EDA), diethylenetriamine (DETA), triethylenetetramine (TETA) and diethylamine (DEA). The adsorption behavior of uranyl ions onto the ATAs was studied in batch experiments. The effects of the contact time, initial concentration of the ions, temperature, and pH value. The salinity were investigated along with the adsorption kinetics and the adsorption isotherms. The kinetic experimental data followed the pseudo second-order kinetic model, and the adsorption isotherms correlated well with the Langmuir model. The ATAs showed good efficiency in adsorbing uranyl ions, with the best saturation adsorption capacity being 64.26 mg g^{-1} for ATA-DETA within 120 min. The temperature dependence of ATA-DETA was quite abnormal and the quickest behavior was obtained at 25°C . ATAs showed good adsorption capacity over a wide pH range of 4.0–8.5, and HCl could be used in the elution process. Salinity of the solution had great effect on the adsorption capacity, 3.5% salinity resulted in a 55% loss of capacity from ATA-DETA. The selectivity of ATA-DETA showed an order of: $\text{UO}_2^{2+} \approx \text{Fe}^{3+} > \text{Zn}^{2+} > \text{VO}_3^- > \text{Co}^{2+} > \text{Ni}^{2+}$.

Keywords: Amino-type adsorbents, Radiation induced graft polymerization, Uranyl ions, Adsorption, Selectivity

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I. INTRODUCTION

Uranium, an important resource of nuclear fuel, mainly resides as sedimentate in terrestrial ores or as dissolved uranyl ions in seawater. The total amount of uranium in seawater is estimated at 4.5 billion tons, about 1000 times the total amount in terrestrial ores, and should be considered as an important fuel source for nuclear power industry [1–3]. However, the low concentration of uranyl ions in seawater, about $3.3 \mu\text{g L}^{-1}$, is a significant challenge in the developing an economic recovery technique.

In the past decades, various methods, such as solvent extraction, flotation, ion-exchange and adsorption etc, have been applied for the recovery of uranium from seawater [4–6]. As a high efficiency and low cost method, the adsorption of seawater uranyl ions with various types of adsorbents has been widely studied and reported.

Of all the adsorbents for uranyl ions coordination reported so far, adsorbents functionalized with amidoxime $[\text{AO}; -\text{C}(\text{NOH})-\text{NH}_2]$ groups are considered as the most promising materials due to their high capacity and selectiv-

ity [7, 8]. Radiation-induced graft copolymerization (RIGC) of acrylonitrile onto polymeric fabric or fibers, and then amidoximed, is now a sophisticated technique in Japan [9]. A marine experiment using a braid adsorbent reported a loading capacity of 2 mg g^{-1} (uranium per adsorbent) [10]. Efforts have been made at SINAP in preparing amidoxime-type adsorbent and studying adsorption behavior of the uranyl ions onto the adsorbent [11, 12].

Amines are widely used to form coordination compounds with metal ions. Therefore, an amino-type adsorbent (ATA) is a worthy candidate for studies on uranium extraction from seawater. Several ATAs utilizing RIGC were reported. The precursor monomer containing an epoxy group such as glycidylmethacrylate (GMA) was grafted onto the polymeric matrix and then amino groups were introduced through a ring-opening reaction [13]. Recently, 4-hydroxybutyl acrylate glycidylether (HB) was applied to synthesize metal ion adsorbents, as it contains an epoxy group too, and due to the longer side chain in HB, the adsorbents performed nicely, with higher adsorption amount and faster adsorption rate than those using GMA as the precursor [13].

In this work, ATAs were prepared by grafting HB onto a non-woven fabric made of polyethylene-coated polypropylene (PE/PP) duplex fiber, and modified with amines of ethylenediamine (EDA), diethylenetriamine (DETA), triethylenetetramine (TETA) and diethylamine (DEA). The adsorption behavior of the uranyl ions onto the ATAs was investigated.

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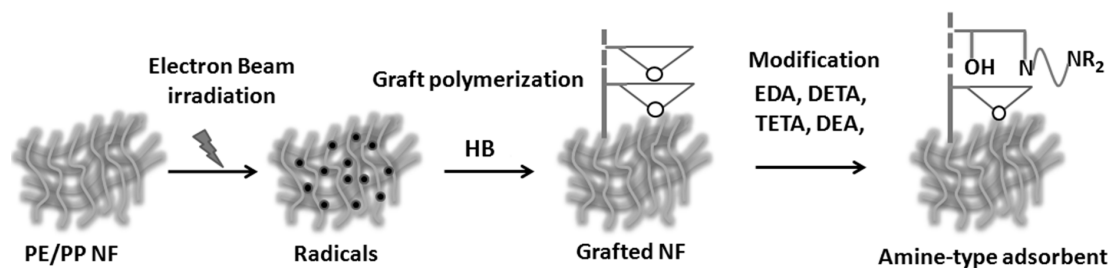


Fig. 1. Preparation of the amine-type adsorbents.

II. EXPERIMENTAL SECTION

Materials: PE/PP duplex fiber non-woven fabric was obtained from Kurashiki MFG Company, Japan. HB was purchased from Tokyo Kasei Kogyo Co. Ltd, Japan. EDA, DETA, TETA, DEA, surfactant sorbitan monolaurate (Span-20), xylene and isopropyl alcohol, were purchased from Kanto Chemical Co., Ltd. Standard solutions of uranyl, iron, vanadium, nickel, cobalt and zinc were bought from Analytical Laboratory, Beijing Research Institute of Uranium Geology. HNO_3 and Na_2CO_3 were purchased from Sinopharm Chemical Reagent Co., Ltd., All the materials and reagents were used without further purification.

Preparation of the ATA: ATAs were prepared using pre-irradiation-induced graft copolymerization and an amination procedure (Fig. 1). The non-woven fabric samples were cooled in dry ice and irradiated 30 kGy by 1.5 MeV electron beam. Graft copolymerization was carried out in an emulsion system containing 5% HB and 0.5% surfactant Span-20 at 40 °C for 2 hours. The degree of grafting (DG) of the obtained fabrics, named as PE/PP-g-PHB, was about 150%. Then, the fabrics were aminated by immersing them in an isopropyl alcohol solution added with EDA, DETA, or TETA (to 70% concentration for each agent), at 60 °C for 4 h. The amination using DEA was done in a 50% DEA water solution at 30 °C for 5 h. The resulting ATAs were named ATA-EDA, ATA-DETA, ATA-TETA and ATA-DEA according to the amine used. The amino group density (AGD) of PE/PP-g-PHB fabrics grafted with the EDA, DETA, TETA and DEA were 3.0 mmol g⁻¹, 1.7 mmol g⁻¹, 1.6 mmol g⁻¹ and 2.2 mmol g⁻¹-adsorbent, respectively.

The AGD was estimated by:

$$\text{AGD} = (Z_f - Z_0)/M/Z_f \times 1000, \quad (1)$$

where, Z_0 and Z_f are the weights of grafted fabric before and after amination and M the molecular weight of the amine compound.

Batch adsorption experiments: To study the adsorption kinetics, ATAs were put into a uranyl ion solution adjusted at pH 8 and kept at 25 °C. The samples were taken out for evaluation at 30 min, 60 min, 120 min, 240 min and 480 min. Once the best ATA was determined, the ATA-DETA adsorbent was used to study the influences of the adsorption. Equilibrium isotherms were determined by varying initial uranium concentration from 1 mg L⁻¹ to 7 mg L⁻¹ and using an equilibrium

time of 8 h. The effect of temperature on the equilibrium uptake of uranyl ions was investigated under similar experimental conditions, except for changes in temperature from 5 °C to 35 °C. The influence of the initial solution pH on uranyl ions adsorption was studied with an initial concentration of 1 mg L⁻¹ and pH value was adjusted between 2.0 and 10.0 using HNO_3 and Na_2CO_3 solutions at 25 °C. Selective metal ions adsorption of uranyl, vanadium, iron, nickel, zinc and cobalt ions onto DETA-type adsorbent tests were performed by using a mixture of an aqueous solution of 1 mg L⁻¹ for each of the six metal ions. The solutions for the batch adsorption experiments were stirred at the same rate at different temperatures for various contact times in a water bath. The concentrations of uranium were determined by a trace uranium analyzer (WJG-III). Selective metal ion concentrations after adsorption were determined by using an ICP (Inductively Coupled Plasma) analyzer (NexION 300 D). The adsorption amount of the uranium ion was calculated using Eq. (2):

$$Q = \frac{1}{238} \frac{(C_0 - C_t)V}{1000w}, \quad (2)$$

where Q (mol g⁻¹) is the adsorption amount of uranium onto amine-type materials, C_0 (mg L⁻¹) and C_t (mg L⁻¹) are the initial concentrations of the uranium in the solution before and after adsorption for a certain time t , respectively. V (L) is the volume of the solution, and w (g) is the weight of the dry adsorbent.

Characterizations: Infrared spectra were taken on a Tensor 27 FT-IR spectrometer (Germany). The pristine and grafted PE/PP duplex fiber non-woven fabrics are scanned in the wave number range of 4000–800 cm⁻¹. Scanning electron microscopy (SEM) images of the pristine PE/PP and ATA-DETA were taken on a JSM-6700F scanning electron microscope (JEOL, Japan). Prior to the SEM observation, the carbon tape for sample attachment were sputtered with gold to enhance the electronic conductivity in vacuum.

III. RESULTS AND DISCUSSION

A. Adsorbent characterization

The HB-graft non-woven fabric and the ATAs were characterized by FT-IR. Fig. 2 shows typical FI-IR spectra of the ATA-DETA, the trunk polymer and HB-grafted non-woven fabric. The spectrum of HB-grafted sample exhibits

strong absorption at about 1730 cm^{-1} (C=O stretching) and 1251 cm^{-1} (—C—O—C stretching), and the peak at 848 cm^{-1} represented the characteristic vibrations of epoxy groups. All these indicate that the HB-graft chains are introduced onto the PE/PP fabric. The ATA-DETA spectrum shows the new characteristic peaks at 1650 cm^{-1} and 1555 cm^{-1} (N—H bending vibration), indicating the successful modification of the DETA group.

SEM images of the virgin PE/PP fabric and ATA-DETA are shown in Fig. 3. The surface image of the ATA-DETA revealed the fiber morphology, crisscrossed by a network of fibers of about $20\text{ }\mu\text{m}$ in diameter (Fig. 3(b)). The rough fiber surface (Fig. 3(c)) means that the ATAs possess an appropriately high specific surface area, which facilitates the adsorption process of uranyl ions.

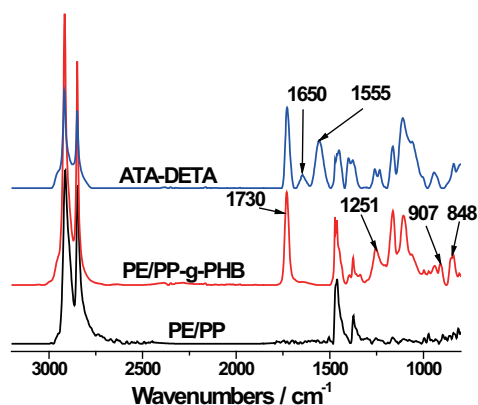


Fig. 2. (Color online) FT-IR spectra of trunk PE/PP non-woven fabric, HB-grafted non-woven fabric and ATA-DETA.

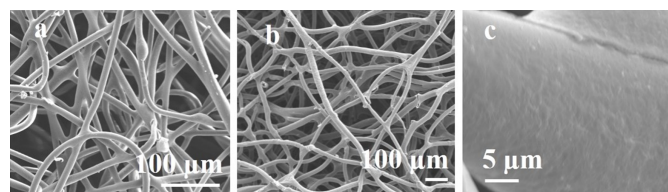


Fig. 3. SEM images of PE/PP (a) and ATA-DETA at low (b) and high (c) magnification.

B. Effect of contact time

The effect of contact time on uranyl ions absorption onto the ATAs was investigated over the time intervals (at 30 min, 60 min, 120 min, 240 min and 480 min). As shown in Fig. 4(a), the amount of adsorption increased with the contact time and finally reached 12.45 mg g^{-1} , 12.50 mg g^{-1} , 12.48 mg g^{-1} and 12.50 mg g^{-1} for ATA-EDA, ATA-DETA, ATA-TETA and

ATA-DEA, respectively. After absorption equilibrium, the four ATAs exhibited similar adsorption amount but the time dependence curves show that the adsorption rate is in the order of ATA-DETA > ATA-DEA > ATA-TETA > ATA-EDA. This means that ATA-DETA exhibited the most efficient adsorption rate for uranyl ions, especially within the first 30 min. As mentioned previously the functional group density of ATAs were 3.0 mmol g^{-1} , 1.7 mmol g^{-1} , 1.6 mmol g^{-1} and 2.2 mmol g^{-1} for ATA-EDA, ATA-DETA, ATA-TETA and ATA-DEA, respectively. Functional groups of the first three type adsorbents were in the form of secondary and tertiary amine when used in aqueous solution and ATA-DEA was of quaternary amine. The most efficient adsorbent in this study was ATA-DETA with a functional group density of 1.7 mmol g^{-1} . Therefore, the adsorption rate of the materials was affected not only by the density of the amino groups, but also the structure of the amino group.

A good correlation with adsorption kinetics data can fully explain the adsorption process. The pseudo second-order model in Eq. (3), which is based on adsorption ability on the solid phase, was used to fit the results obtained from the different amine-type materials.

$$\frac{t}{Q_t} = \frac{1}{kQ_e^2} + \frac{t}{Q_e} \quad (3)$$

where k is the rate constants of the pseudo second-order adsorption, Q_t and Q_e are the amount of adsorbed uranium at time t and equilibrium state. The fitting results applying the pseudo second-order model to the kinetic data of different ATAs is given in Fig. 4(b), and one sees a good linearity with the correlation coefficient ($R^2 > 0.99$) under the experiment conditions.

As ATA-DETA has the most efficient adsorption rate for uranyl ions, we focused on ATA-DETA in the following experiments.

C. Effect of the initial concentrations

The effect of initial uranyl ion concentration on the ATA-DETA adsorption was obtained from 1 to 7 mg L^{-1} at a pH 8.0 at $25\text{ }^\circ\text{C}$. Fig. 5 shows the amount of uranyl ions adsorbed on the ATA-DETA om 8 hours. The amount of adsorbed uranyl ion increases with the initial concentrations and reached a plateau value of 64.26 mg g^{-1} , i.e. the maximum equilibrium adsorption amount.

The Freundlich and Langmuir adsorption models are two typical adsorption equilibrium isotherms widely used to describe adsorption equilibrium. The Freundlich model supposes that a nonideal adsorption surface is heterogeneous with multilayer sorption [14]. The linear form of the Freundlich model is given by

$$\log Q_e = \log K_F + \frac{1}{n} \log C_e, \quad (4)$$

where K_F and n are the Freundlich constants to indicate the adsorption capacity and the extent of the adsorption, respectively [15]. Q_e (mg g^{-1}) is the amount of solute adsorbed at

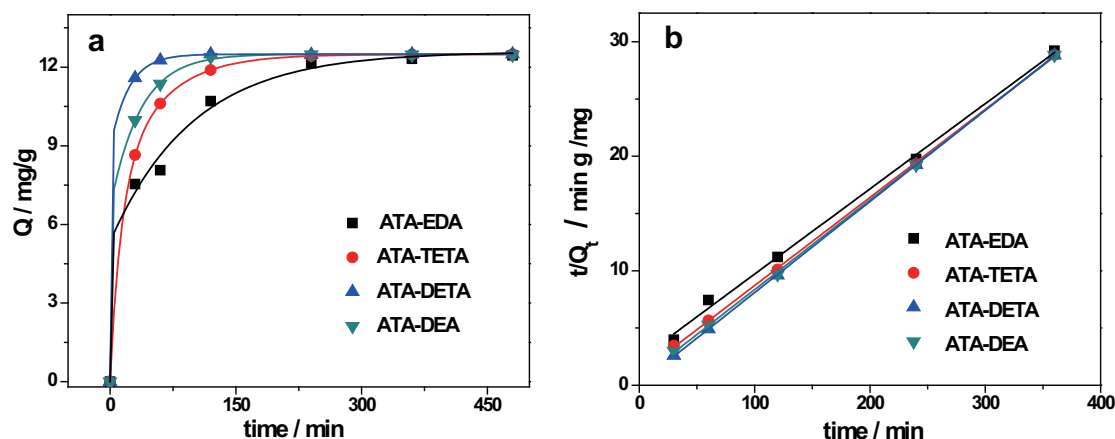


Fig. 4. (Color online) (a) Adsorption kinetics of different ATAs ($C_0 = 1 \text{ mg L}^{-1}$; $T = 25^\circ \text{C}$; $\text{pH} = 8$) (b) Linearized pseudo-second-order kinetic model for uranium on different ATAs.

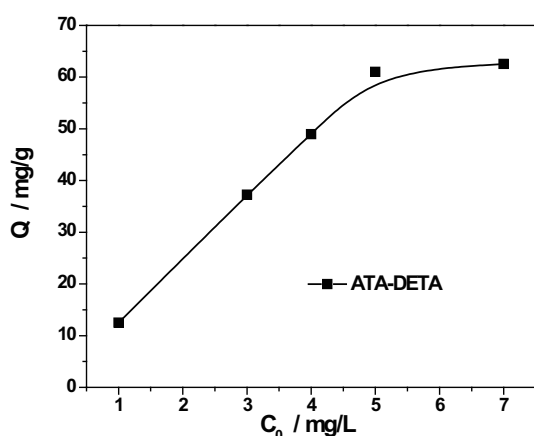


Fig. 5. Effecting of the initial concentration on adsorption capacity.

equilibrium, and C_e (mmol L^{-1}) is related to the concentration equilibrium.

The Langmuir model assumes that the adsorption of metal ions occurs on a homogeneous surface by monolayer adsorption without any interaction between adsorbed ions [16]. The linear form of the Langmuir model is given by:

$$\frac{C_e}{Q_e} = \frac{C_e}{Q_{\max}} + \frac{1}{Q_{\max} K_L}, \quad (5)$$

where, Q_{\max} is the Langmuir monolayer adsorption capacity (mg g^{-1}) and K_L is the Langmuir equilibrium constant: the ratio of adsorption and desorption rate coefficients (L g^{-1}).

The plots based on the two linear relationships are shown in Fig. 6. The data were linearly correlated, with the correlation coefficients (R^2) of 0.877 and 0.999 for the Freundlich and Langmuir models, respectively. This indicates that isotherm is a better fitting model than Freundlich and the adsorption of

ATAs for uranyl ions occurred on a homogeneous surface by monolayer adsorption.

D. Effect of the temperature

Temperature is an important parameter for ion adsorption. In the early marine experiments in Japan, scientists found materials set in Okinawa showed higher adsorption capacity than that in the north Japan, indicating that high temperature took advantage for uranyl ion adsorption. It was also reported that higher temperature resulted in higher uranyl ion adsorbance for the amidoxime-type adsorbent in laboratory study [11]. These suggest that the adsorption procedure maybe endothermic. However, in this study, 25°C was found to be the most optimal temperature for the adsorption of uranyl ions of ATA-DETA. The effect of the temperature on uranyl ions adsorption onto ATA-DETA is given in Fig. 7(a). The pseudo second-order curves were used to fit the data obtained at different temperatures for the ATA-DETA adsorption, with good correlation coefficients ($R^2 > 0.98$) (Fig. 7(b)). The adsorption rates were in the order of $25^\circ \text{C} > 35^\circ \text{C} > 15^\circ \text{C} > 5^\circ \text{C}$. The Arrhenius equation was used to study the temperature dependence of the reaction rate constant, but the activation energy (E_a) could not be calculated from this empirical relationship. This indicates the adsorption process involves both endothermic and exothermic in the complex step. As the dominant uranyl species in seawater are the carbonate complexes [17, 18], the amidoxime must compete with and replace the carbonate groups in the sorption process. Literature reports suggest that the dissociation of the tricarbonato uranyl complex ($\text{UO}_2(\text{CO}_3)_3^{4-}$) may be the rate-determining step [19]. However, each step of the dissociation of the tricarbonato was not clear at present, nor the thermodynamics involved in dissociation of each carbonate. To learn more about the adsorption mechanism, a theoretical simulation will be included in our future study.

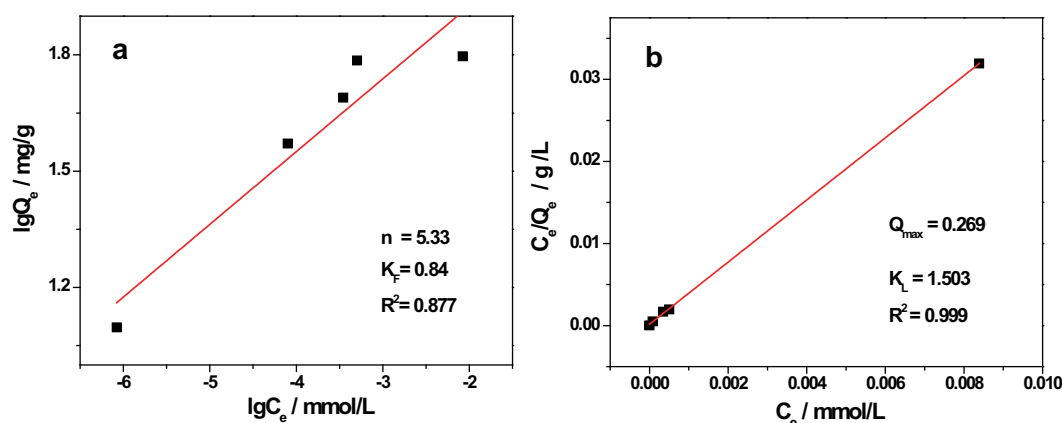


Fig. 6. (Color online) Fitting of uranium adsorption on ATA-DETA ($T = 25\text{ }^{\circ}\text{C}$, $\text{pH} = 8$, $t = 8\text{ h}$) (a) Freundlich adsorption isotherm and (b) Langmuir isotherm.

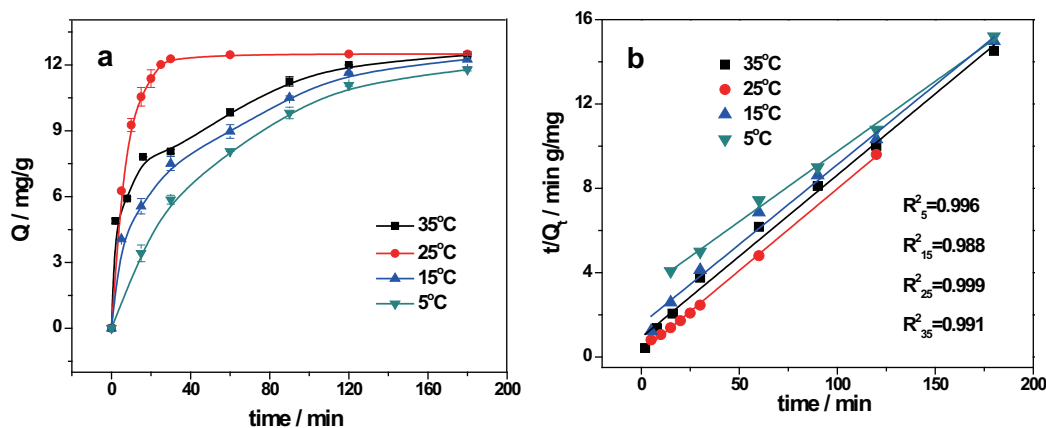


Fig. 7. (Color online) (a) Adsorption kinetics of different temperatures with ATA-DETA ($C_0 = 1\text{ mg L}^{-1}$; $T = 25\text{ }^{\circ}\text{C}$; $\text{pH} = 8$) (b) Linearized pseudo-second-order kinetic curves for uranyl ion adsorption at different temperatures.

E. Effect of the pH value

The pH value plays a role in metal ions absorption because of the hydration and complex formation of metal ions. Although the pH value of the seawater is constant at about 7.5–8.5, looking at the effect of the pH value shall be of help to determine the desorption conditions. The results of pH influence (pH 2.0–9.5) is given in Fig. 8.

In the range of pH 4.0–8.5, the adsorption ratios and amount of the uranyl ions were almost the same, while a pH value over 8.5 resulted in a slight decrease. The adsorption ratio and adsorbance of uranyl ions dramatically decreased in an acid condition with a pH value below 4. At pH 2, the ATA-DETA is incapable of adsorbing the uranyl ions.

The pH effect can be explained in two ways. First, the existing form of $(\text{UO}_2)^{2+}$ in aqueous solutions is extremely complex. U (VI) mostly exists as $(\text{UO}_2)^{2+}$ in its hydrolysis complexes, carbonate complexes and multinuclear hydroxide as a

function of pH and concentration under experimental conditions [18, 20–22]. $(\text{UO}_2)^{2+}$ were the main species in acidic solution at pH 2–4 [23]. At pH 4–8.5, $(\text{UO}_2)^{2+}$, $[\text{UO}_2\text{OH}]^+$, $[(\text{UO}_2)_3\text{O}(\text{OH})_3]^+$, $[\text{UO}_2(\text{CO}_3)]$, and $[(\text{UO}_2)_2(\text{OH})_2]^{2+}$ coexist [19–22]. On the other hand, the protonation of amino groups depends greatly on the pH value of the solution, where the lone pair electrons on N are occupied by hydrogen, which makes coordination of amine absent in strong acid condition. This illustrates that desorption procedure can be carried out in acidic solution.

After adsorption equilibrium in 1 mg L^{-1} uranyl ions solution, desorption was carried out with HCl. The desorption ratio of uranyl ions from ATA-DETA under different concentrations of HCl is shown in Table 1. The results show that 0.1 M HCl can remove 75.11% uranyl ions from ATA-DETA. Higher concentration of HCl is of higher desorption ratio and 5 M HCl can remove 93.67% uranyl ions from ATA-DETA.

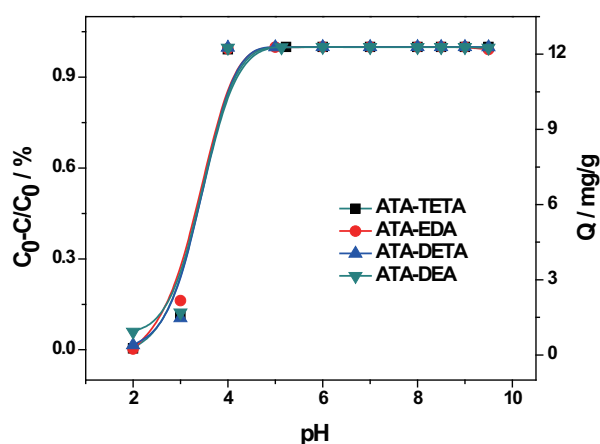


Fig. 8. (Color online) Effect of solution pH on the equilibrium adsorption efficiency onto ATAs ($C_0 = 1 \text{ mg L}^{-1}$; $T = 25^\circ\text{C}$; $t = 24 \text{ h}$).

TABLE 1. Desorption ratio of uranyl ions from ATA-DETA with different concentration of HCl

HCl concentration (mol L^{-1})	0.1	0.5	1	5
Desorption Ratio (%)	75.11	90.02	91.17	93.67

F. Effect of salinity

Salinity is important for the recovery or separation of seawater uranium due to the high average salinity (3.5%) of seawater. Fig. 9 shows the salinity effect on equilibrium adsorption amount.

The salinity was adjusted with NaCl. The adsorption of uranyl ion was affected a little at salinities below 1%, but it decreased quickly with increasing salinity from 1% to 4%, where it began to decrease slowly until 5% salinity, and reached an equilibrium at 6% salinity and 4.76 mg g^{-1} of the adsorption. It has been reported that the salinity had great effect on the adsorption of uranyl ions [15–17]. However, with a 55% loss of capacity from the ATA-DETA at salinity of 3.5%, the salt resistance property of the adsorbents needs to be improved, which is a challenge of adsorbent design.

G. Effect of interfering ions

In a practical application, co-existing ions which would strongly interact with the amino groups will interfere with the adsorption of uranyl ions. Co^{2+} , Fe^{3+} , Zn^{2+} , Ni^{2+} and VO_3^- were investigated in this study as the concentration of Fe^{3+} , Zn^{2+} , Ni^{2+} and VO_3^- and uranyl ions in seawater are at the same level of ppb ($\mu\text{g L}^{-1}$). Moreover, the distribution coefficient of Co^{2+} , Fe^{3+} and Ni^{2+} from the amidoxime-type adsorbents are higher than that of uranyl ions [24]. Vanadium also attracted wide attention recently as it can be collected on the adsorbent, but the vanadium elution is difficult. The adsorption amount of uranyl ions in mix solutions at pH 8 decreased from

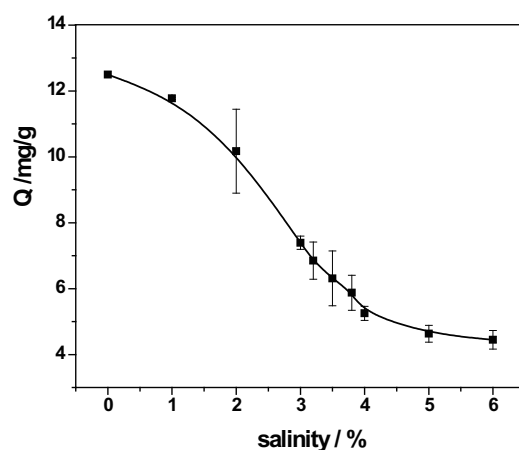


Fig. 9. Salinity effects on the equilibrium adsorption amount. ($C_0 = 1 \text{ mg L}^{-1}$; $T = 25^\circ\text{C}$; $t = 24 \text{ h}$)

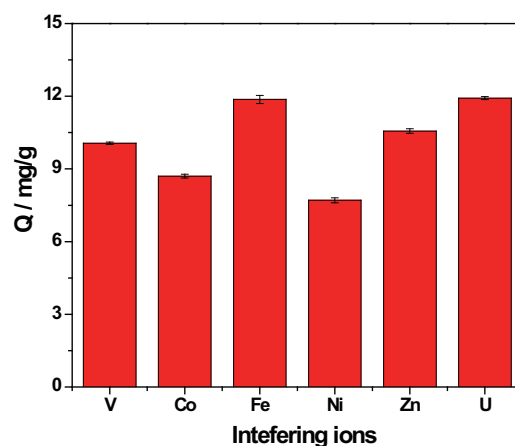


Fig. 10. (Color online) Adsorption amount of different metal ions by ATA-DETA ($C_0 = 1 \text{ mg L}^{-1}$; $T = 25^\circ\text{C}$; $t = 24 \text{ h}$, pH = 8).

12.50 mg g^{-1} to 11.92 mg g^{-1} when the initial concentrations of interference were all set at 1 mg L^{-1} . Fig. 10 indicates that the sequence of adsorption amount of metal ions is: $\text{UO}_2^{2+} \approx \text{Fe}^{3+} > \text{Zn}^{2+} > \text{VO}_3^- > \text{Co}^{2+} > \text{Ni}^{2+}$. The adsorption capacity of uranium was higher than vanadium, which is an advantage over amidoxime-type adsorbents [25]. The interference of Fe^{3+} , however, still presents challenges in the preparation of adsorbents with high selectivity.

IV. CONCLUSION

ATAs exhibit excellent adsorption behavior of uranium with saturation sorption capacities as high as 64.26 mg g^{-1} from the ATA-DETA type adsorbent. The sorption kinetics followed a pseudo second-order kinetic model, while the sorption isotherm followed the Langmuir adsorption isotherm model.

ATAs showed good adsorption capacity over a wide pH range of 4.0–8.5. Using 0.1 M HCl, 75.11% uranyl ion could be removed from the ATA-DETA, and the desorption ratio increased to 93.67% with 5 M HCl. The most suitable temperature for the adsorption of uranyl ions was 25 °C. A salinity

of 3.5‰ resulted in a 55% loss of capacity from ATA-DETA in this study. The selectivity from ATA-DETA was in the order of $\text{UO}_2^{2+} \approx \text{Fe}^{3+} > \text{Zn}^{2+} > \text{VO}_3^- > \text{Co}^{2+} > \text{Ni}^{2+}$. The adsorption capacity of uranium was higher than that of vanadium, which is an advantage over other ATAs. The effect of Fe^{3+} , however, remains a challenge.

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